ADVANTAGES AND DRAWBACKS OF TUBULAR FLOW BURNER FOR TESTING FLAMMABILITY LIMITS

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ABSTRACT

In this work. the tubular flame burner is assessed as a fast and inexpensive tool for determining the flammability limits of hydrocarbon blends with and without suppressants. Ths variation of temperature of the unburned combustion gas was studied. and preheating was found to be unavoidable during burner operation. even when the burner incorporated a cooling jacket. The analysis of flammability limits of two hydrocarbon/air mixtures (natural gas and a refrigerant blend), diluted with nitrogen and carbon dioxide, shows that the preheating of the gas appears to he responsible for the increased flammability range obtained in previous studies on tubular flame burners. The correction procedure involves the calculation of the adiabatic flame temperature and allows the reduction of the natural gas results to methane. for comparison with published data. Once corrected for the effect of preheating. the results obtained from the tubular burner follow closely those derived from the Bureau of Mines apparatus.

The upper flammability limit was observed to depend on the injection velocity of the nitrogen shroud gas, hut only when this velocity was lower than the injection velocity of the unburned mixture. The measured limits of flammability were found to vary with the injection velocity. with a maximum flammable range obtained at a velocity of 5 cm/sec. This velocity minimises the effects of heat losses (which are important below 5 cm/sec) and flame stretch (which becomes significant at hiyher velocities). For the size of porous cylinder used in the present work, migration of the flame towards the cylinder wall (for Le>1 flames at extinction) does not appear to have a noticeable effect on the flammability limit. Overall. we find that the tubular burner provides a more convenient method for determining flammability limits in comparison to the repetitive testing required by ISO 10156 and ASTM E681 standards. On the other hand, the tubular burner consumes larger volumes of the test mixtures.

INTRODUCTION

The development of new gaseous fire suppressants and refrigerants necessitates the determination of fire-extinguishment efficiency of suppressants and flammability properties of refrigerants. These two properties can be quantified by measuring the flammability limits of refrigerants and flammable mixtures doped with fire suppressants. The first experimental method for obtaining flammability limits was developed by Coward and Jones [1], and later reviewed by Zabetakis [2]. This method consists of an explosion burette into which a fuel/oxidiser mixture is introduced. The gas mixture is deemed flammable if a flame successfully propagates the length of the tube after the activation of the ignition source [3]. Although the extensive number of results obtained by this method have been accepted for many years, it has become evident that the results depend on the tube length. its diameter. and the direction of flame propagation. In spite of the fact that a small amount of refrigerant or fire suppressant is needed in each experiment, the method requires repetitive tests to obtain good estimates of flammability limits (or flammability envelope). This

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makes the method expensive and time consuming to operate. For these reasons, we examine herein the applicability of the tubular flow burner for rapid determination of flammability limits.

A study by Hertzberg [4] describes fundamental processes that may complicate the measurement of flammability limits in physical apparatus. These include heat losses from the flame by conduction, convection and radiation to the apparatus walls, instabilities in the flame front resulting from buoyant convection, selective diffusional demixing and flow gradients (flame stretch), as well as radical loss or their generation on apparatus walls. It is now acknowledged that the more significant of these processes are the heat loss from the flame, flame stretch as a result of the fluid dynamics aspect of the flow, and the selective diffusional demixing [5].

In more recent work, it has become desirable to obtain a more fundamental measurement of flammability through the use of an apparatus that minimises heat losses and allows flame stretch and flame-front instabilities to be characterised. This desire came from the realisation that flammability limits are controlled by external (e.g., heat losses or flame stretch) and internal (e.g., preferential diffusion of a limiting component) parameters [5]. The effect of external parameters needs to be minimised or controlled to obtain extinction data of a more fundamental nature, which are independent of a testing apparatus. This has led to the development of new methods for measurement of flammability limits.

One such method devised to factor out the effect of flame stretch is that used by Womeldorf and Grosshandler [6] in an apparatus that creates two identical, opposed-flow, premixed gas streams. Nearly adiabatic twin flames are formed, with their stretch rates controlled by adjusting the flow rates of the gas streams. The stretch independent value of the lean flammability limit is obtained by extrapolating the fuel equivalence ratio versus the global strain rate to zero stretch. The flammability measurements obtained by this system are similar to those collected from ISO 10156 and **ASTM** E681 [7] tests, but in addition carry more fundamental meaning since they reflect the data [3] corresponding to the limit of vanishing flame stretch.

In yet another attempt to obtain a more fundamental measurement of flammability, Ishizuka [5] developed an apparatus, which produces a tubular flame of circular cross section, existing in a stretched flow field, inside a porous tube. Downstream heat losses are considered to he negligible due to the flame existing in a counter flow field with axial symmetry. In addition, lateral conductive losses are minimised due to the ends of the tubular flame having a smaller area than in the case of twin flames. Increased flammability ranges were obtained using a tubular flame burner in numerous investigations [e.g., 8,9], and these results were directly attributed to the reduction in downstream heat losses. However, the data presented in this paper appear to refute this explanation.

The tubular flame burner was used to determine flammability limits of natural gas and refrigerant blend with and without nitrogen and carbon dioxide diluents. The results are then compared with those previously obtained by others from the tubular flame burner and the Bureau of Mines apparatus. The effects of preheating as well as injection velocities of unburned gases and nitrogen shroud, on flammability limits were also investigated. This was done to determine whether the tubular flame burner provides reproducible and reliable extinction results that compare well with the standard ISO and ASTM tests. It was concluded that the tubular flame burner yields rapid measurements of flammability limits, and is especially useful when a large number of suppressants need to be tested.

EXPERIMENTAL APPARATUS

A detailed diagram of the tubular burner is shown in Figure 1. The burner consists of a porous brass cylinder with identical dimensions to those used in previous studies [5,8,9]. The brass cylinder is 30 mm in inside diameter and is 80 mm in length. The cylinder wall is 5 mm in thickness and has pores 5 µm in diameter. The combustible gas mixture is passed into the jacket surrounding the brass tube and proceeds in a radial direction through the pores and toward the internal cylinder axis. When ignited, *an* axisymmetric tubular flame of circular cross section is formed in a stretched flow field. The burner is operated in the vertical orientation, although a horizontal orientation is also possible [5].

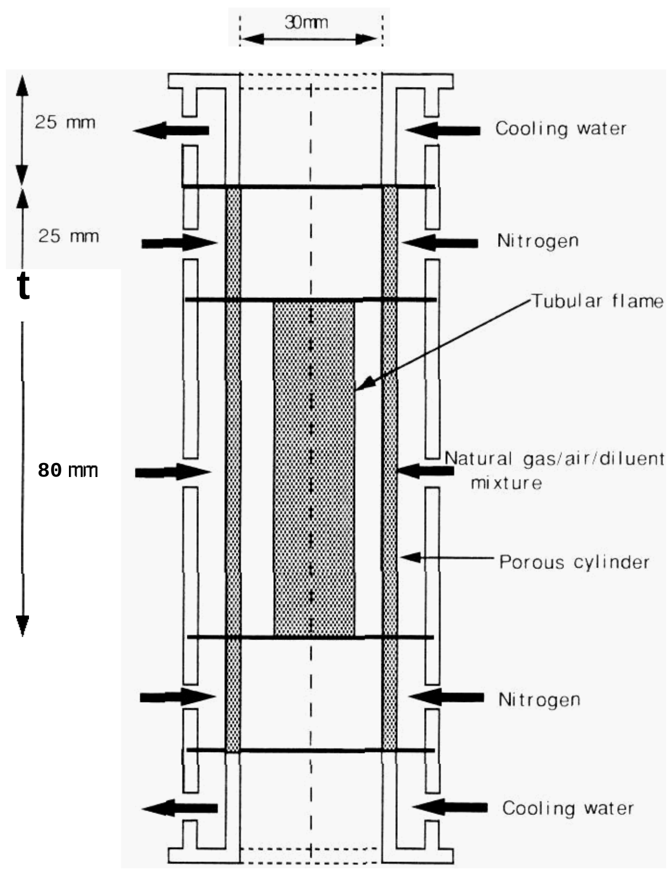

Figure 1. Detailed diagram of the tubular flame burner.

The burner is equipped with a nitrogen injection section of 25 mm in length, to quench and extinguish the flames. Nitrogen is injected with a velocity equal to the velocity of the combustion mixture. This injection was especially important close to the upper flammability limit, in order to prevent the formation of diffusion flames inside and above the burner. Two 25 mm in length water cooling ends are located on each side of the nitrogen injection sections to provide cooling of the burner.

Figure 2 illustrates the tubular flame burner and gas flow system used in this investigation. The air is supplied at pressure via a compressor and metered by a rotameter. Fuel and suppressant flows are controlled by mass-flow controllers, which are calibrated with a soap bubble meter. An exhaust vent is located above the burner to remove any hazardous products of combustion safely.

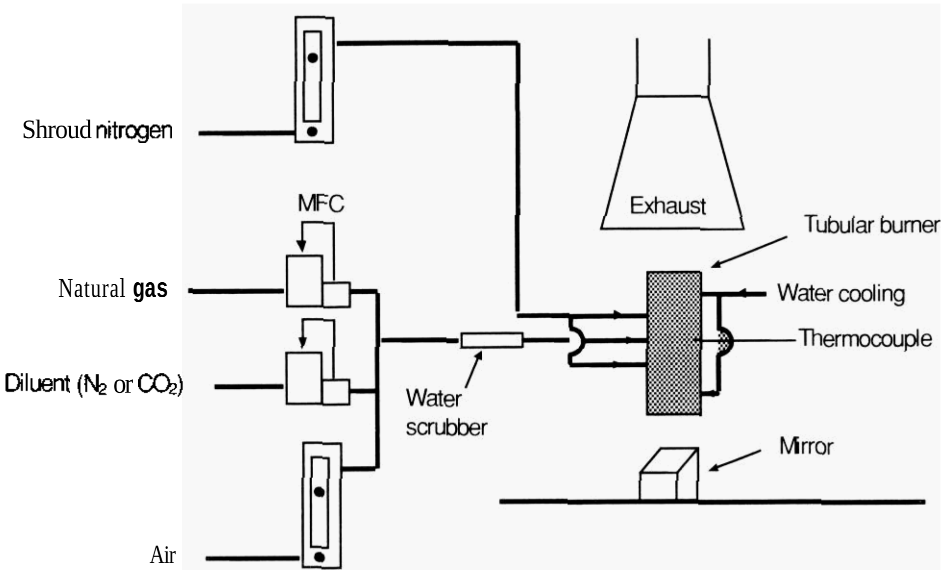

Figure 2. Diagram of tubular flame burner apparatus (MFC = mass flow controller).

A thermocouple was inserted into the jacket surrounding the porous tube to measure the unburned gas temperature. The temperature of the unburned gas affects the flammability limits, and this modification was needed to verify and quantify the effect the unburned gas temperature on the measured flammability limits.

In the first step of the experimental procedure, the burner was supplied with a combustible gas mixture, which was ignited with an electric spark. A small amount of time was allowed for the flame to stabilise and assume a uniform cylindrical shape. The flammability limits were determined by altering the fuel concentration of the mixture stepwise until the flame could no longer be sustained. A constant injection velocity was maintained throughout each experiment. Peak concentrations of suppressants were measured by igniting a flammable mixture and then approaching the peak concentration region by gradually increasing the concentration of the suppressant until extinguishment occurred. In all experiments, a mixture was judged to be flammable if the flame was sustained for at least 1 min.

RESULTS

The goal in this study was to determine whether the tubular flame burner could serve as a fast, inexpensive, and easy to operate testing tool for the determination of flammability limits. For this reason, investigations were conducted of the effects of the shroud gas injection, unburned gas temperature and its injection velocity, as well as concentration of inert suppressants (N_2 and CO_2, both >99% purity) on the flammability of two hydrocarbon mixtures. The analyses of these two hydrocarbon mixtures are listed in Table 1.

TABLE I. FUEL COMPOSITION FROM GAS CHROMATOGRAPHIC ANALYSIS.

	Species	Composition, %
Natural gas	Methane	84.7
	Ethane	12.3
	Carbon dioxide	2.3
	Nitrogen	0.7
Blend **A**	Propane	65.9
	lsobutane	32.0
	Butane	1.5
	Ethane	

Effect of Nitrogen Injection Velocity on Fuel Rich Limit

To prevent the formation of diffusion flames for rich limit mixtures. nitrogen gas was injected into each end of the burner. The effect of the nitrogen injection on the upper limit of blend A (Table I) was investigated and results can be seen in Figure 3. It is clear that the rich flammability limit tends to decrease and then level off as the nitrogen injection velocity approaches that of the combustion mixture (5 cm/sec).

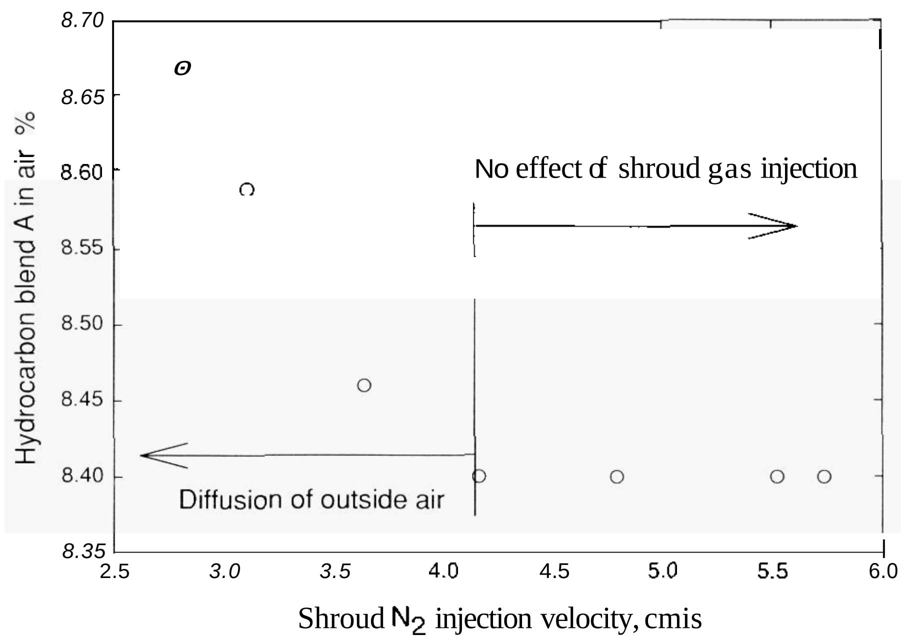

Figure 3. Effect of nitrogen injection velocity on fuel rich limit; injection velocity of the hydrocarbon blend A is 5 cm/sec. The blend contains 0.6% of ethane, 65.9% of propane, 1.5% of butane and 32% of isobutane (Table I).

At low nitrogen injection velocities, *a* bright diffusion flame was observed within the tubular flame, corresponding to oxygen from the atmosphere diffusing into the combustion zone and reacting with the excess fuel. This excess oxygen allows flames of greater fuel concentration to stabilise in the burner, as a result of the increased amount of energy released. **As** the nitrogen velocity increases, the measured upper limit decreases due to more efficient suppression until a constant limit is obtained. Based on this result, a nitrogen injection velocity equal to that of the combustion mixture was selected for subsequent experimentation. This is in agreement with the previous studies [5,8,9].

Effect of Unburned Gas Temperature

In the preliminary experiments, it was observed that significant heating of the burner took place during operation. It was evident that some degree of preheating of the combustion mixture would result and therefore influence the limits measured. Figure 4 illustrates the effect of unburned gas temperature on the lean flammability limit of natural gas/air mixture. The temperature range covered is from 70 to 140 %. After the gas mixture was ignited and sufficient time was allowed for the flame to stabilise, it was found that the minimum unburned gas temperature was approximately 70 "C, when the first limit composition was reached. This result confirms that preheating of unburned gas mixture is inherent to the operation of the tubular flame burner. This factor is unaccounted for in previous studies [5,9]. By altering the amount of time taken to reach the lean limit mixture, the unburned gas temperature was increased and limits were measured.

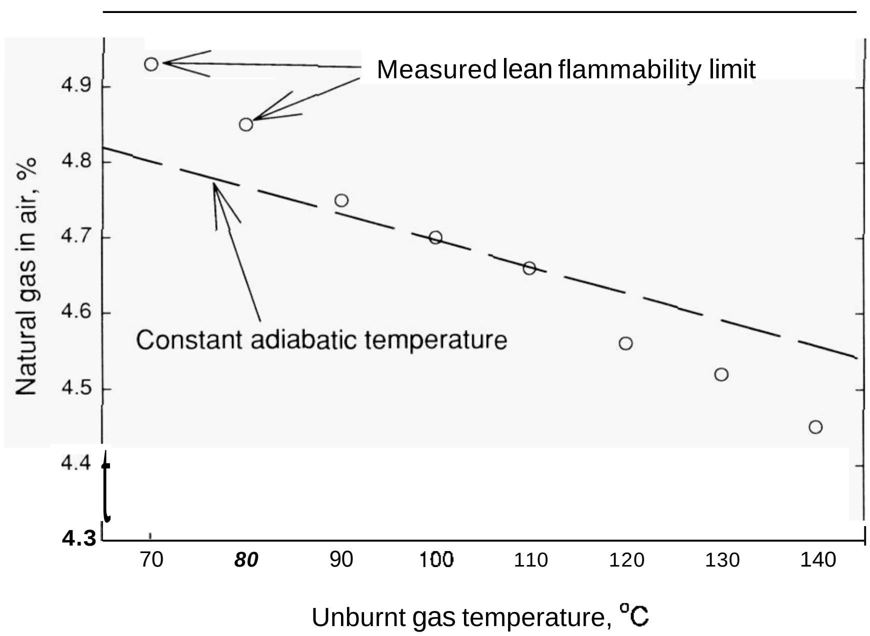

Figure 4. Effect of unburned gas temperature on the lean limit of natural gas; the injection velocity of natural gas is 5 cm/sec.

The lean flammability limit of natural gas (Figure 4), can be seen to vary by approximately *0S%* over the temperature range covered. This result suggests that the preheating of the combustion mixture is a valid reason for the increased flammability range reported [5,8,9] in previous studies using a tubular flame burner.

Figure 4 also illustrates that flames of lower fuel composition can be sustained in the burner as the degree of preheating is increased. This is in agreement with the existence of a critical flame temperature corresponding to the lean flammability limit [e.g., 10]. Due to the increased energy provided on preheating, a leaner fuel mixture reaches the critical limit temperature. **An** average adiabatic flame temperature of 1539K was obtained at the lean limit for the natural gas/air mixtures, which is in good agreement with the accepted value of around 1500 K for short chain hydrocarbons. The dashed line in Figure 4 represents the variation of fuel/air composition for a constant adiabatic flame temperature of 1539 K. Extrapolating to standard conditions (25 °C), a lean limit of approximately *5%* natural gas is obtained, which ***was*** expected since the major component of the natural gas was methane. The deviation of the data points from the dashed line can be attributed to velocity effects due to the preheating of the gas. The mixture injection velocity **was** maintained at 5 cm/sec measured at 25 °C; however, this would change in situ because of the expansion of the gases with preheating.

Effect of Combustion Mixture Injection Velocity

The response of the measured flammability limits at different injection velocities for natural gas/air and hydrocarbon blend A/air mixtures is shown in Figure 5, where it is seen that the flammability ranges both for natural gas and blend **A** are increased as the injection velocity approaches 4.5-5 cm/sec, with the maximum flammable ranges occurring at approximately 5 cm/sec. This value is in direct agreement with the results of Ishizuka [5], where the widest range of flammability was obtained at the injection velocity of 5 cm/sec. The widening of the flammability limits with increasing velocity was directly linked to the visual observations made regarding the flamc geometry and behaviour at extinction.

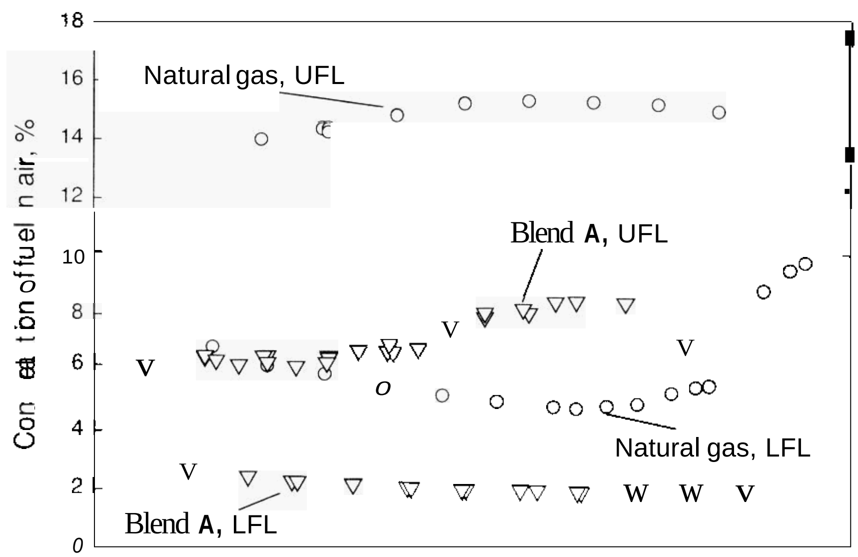

Figure 5. Effect of combustion gas injection velocity on flammability limits of natural gas and hydrocarbon blend **A;** the temperature of the unburned gas is around 80 °C.

For the methane flames. at very low injection velocities, the flames remain close to the burner tube. This condition facilitates losses of heat and radical species to the wall. On approaching the extinction composition. the flame becomes unstable and oscillates. resulting in a disturbed flow field, as also observed by Ishizuka [5]. The combination of these factors is the cause of the decreased flammability range. On increasing the injection velocity from the minimum value, a subsequent increase in flame stability is witnessed, as the flame remains completely stable when approaching the limit composition for injection velocities in excess of 4 cm/sec.

The narrowing of the natural gas/air flame was more pronounced at the lean limit with the flame forming an almost "solid" rod in the centre of the burner. This result corresponds to the lean mixture having a value of $Le<1$ [5] where significant narrowing of the tubular flame occurred with mixtures of this kind. The Lewis number compares thermal diffusivity of the gas mixture with the diffusion coefficient of the deficient species, and is an indication of the relative rates of heat and mass transfer. The Lewis number of less than unity implies faster diffusion of the deficient species in the flame than is the case for heat conduction. It can also be seen that the variation in the lower flammability limit is more pronounced than in the rich limit (Figure 5). These results would appear to follow from the reduction in heat losses and migration of radical species to the burner wall due to the greater degree of narrowing of the tubular flame when $Le<1$.

For injection velocities above 5 cm/sec, it can be seen that the measured limits begin to narrow very slightly as the velocity is increased. This small decrease in the flammability range is a direct result of the increasing flame stretch rate. In the case of natural gas/air flames, a sharp rise was obtained for injection velocities above 5.7 cm/sec, which was associated with the observed flame instabilities. This result was inconsistent with those obtained by Ishizuka [5] who observed a very small and gradual increase in the lower flammability limit up to approximately 20 cm/sec.

With respect to the hydrocarbon blend **A,** it is the upper flammability limit that shows more variation than the lean limit. This can be attributed, similarly to the LFL of natural gas/air mixtures, to $Le<1$ of fuel rich mixtures of blend **A** and air. For injection velocities in excess of 4.5 cm/sec, the flame increases in stability near the limit composition and forms a multipetal pattern similar to that described by Ishizuka [5].

Flammability of Natural Gas/Air/Diluent Mixtures

Figure 6 contains the measured limits for methane/air mixtures suppressed with carbon dioxide and nitrogen. The limits were obtained by using natural gas and correcting the data points to methane. This was achieved by determining the methane composition corresponding to the calculated adiabatic flame temperature for each experimental data point. Thermodynamic calculations were performed using the equilibrium (Equilib) program included with the Chemkin distribution.

The results reported in Figure 6 were obtained for the injection velocity of **5** cm/sec and an unburned gas temperature of 80 ± 2 "C. Also shown in Figure **6** are the results obtained by Liao et al. [9] using tubular burner and Coward and Jones [1] using the Bureau of Mines apparatus. The Bureau of Mines data were measured with an unburned gas having the temperature of 25 °C, whereas the tubular flame data of Liao et al. were reported without temperature measurement. **As** can be seen in Figure 6, the flammability ranges measured using the tubular flame burner are greater than the Bureau of Mines data for both nitrogen and carbon dioxide diluents. The peak

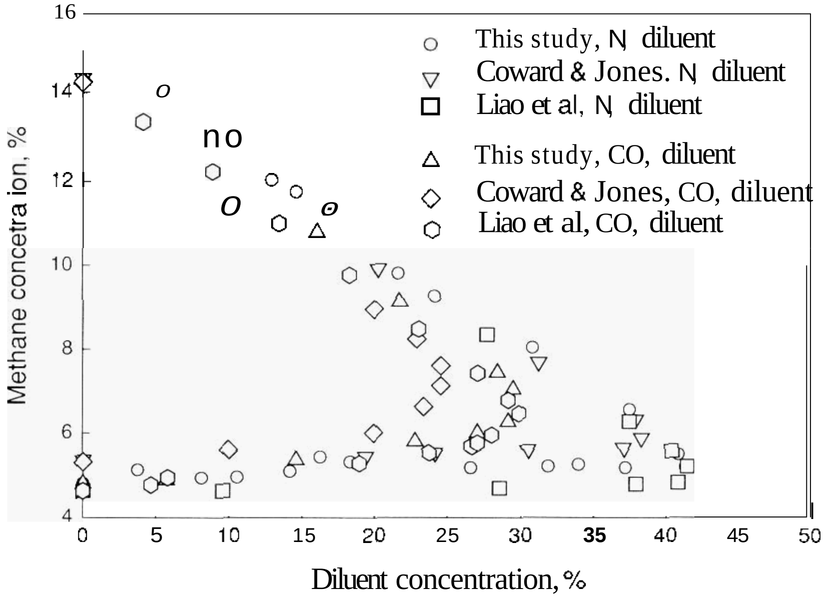

Figure 6. Flammability of natural gas/air/diluent mixtures; the natural gas concentratinn was corrected to methane through the calculation of the adiabatic flame temperature.

concentrations determined in this investigation for both nitrogen and carbon dioxide were found to be significantly greater than in the Bureau of Mines data, and agreed accurately with the results of Liao et al [9]. This is further illustrated in Table 2, which lists the upper and lower flammability limits, and peak concentrations for both nitrogen and carbon dioxide. from these three studies. The results of the Bureau of Mines apparatus clearly indicate a lower range of flammability: however, this outcome was expected due to the preheating of the combustion gas experienced in this investigation and presumably in the Liao et al. measurements.

TABLE 2. COMPARISON OF FLAMMABILITY DATA-
ALL RESULTS REPORTED IN VOL.%.

	Present work (T=80 °C)	Liao et al. [9] (T unknown)	Coward and **Jones** [1] (T=25 °C)
Upper Flammability Limit	15.3	15.4	13.8
Lower Flammability Limit	4.7	4.7	5.3
Peak Concentration N_2	41.4	41.5	38.4
Peak Concentration CO_2	29.2	29.7	24.7

To make **a** better comparison between the results obtained in this investigation and the Bureau of Mines data, thermodynamic calculations were performed to correct the unburned gas temperature from 80 to 25 "C. This was again achieved using the equilibrium program from the Chcmkin distribution. An adiabatic flame temperature was calculated for each data point with an initial temperature of 80 "C. **A** methane composition was then calculated so that an equivalent adiabatic flame temperature **was** achieved. however, this timc with an initial temperature of 25 "C. The results are seen in Figure 7.

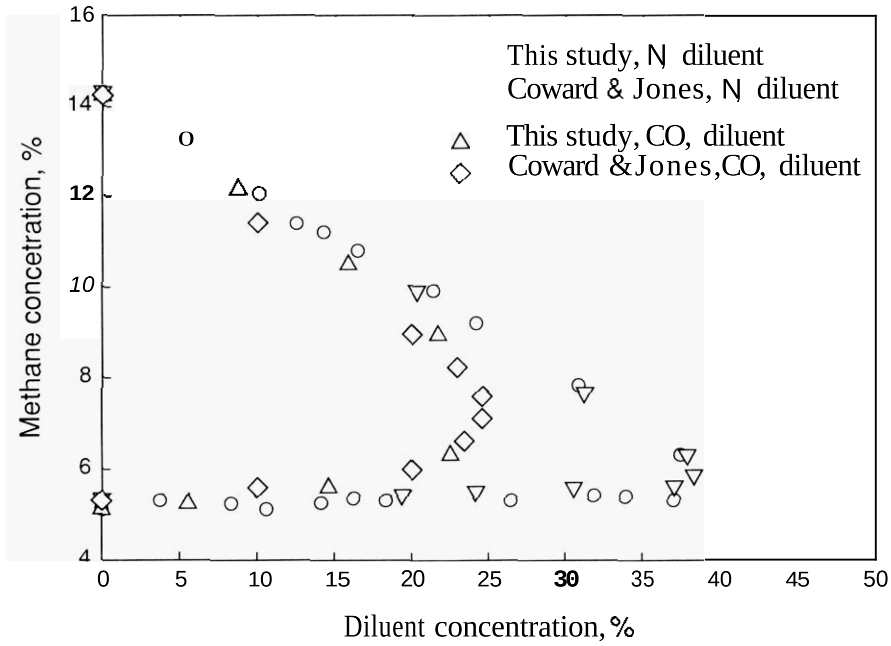

Figure 7. Flammability of methane/air/diluent mixtures corrected to 25 "C.

Figure 7 demonstrates a very good agreement between the tubular flame data of this study and the Bureau of Mines results after the gas preheating is taken into account. Based on this result, it is evident that the increased flammability ranges obtained in previous studies [e.g., 9] would he at least partly due to preheating of the combustion gas, and not entirely a result of a reduction in lateral conductive heat losses in comparison to twin flame geometry, as surmised by Ishizuka [5]. This indicates that a thermocouple is a necessary addition to the tubular flame burner system, for accurate measurement of the temperature of the unburned gases.

CONCLUSIONS

In this study it has been demonstrated that the tubular flame burner provides repeatable and reliable extinction data, which are in agreement with the results of the previous measurements. **A** burner **30** mm in diameter is sufficiently large in size to minimise the effects of Le>1, when the flame migrates towards the porous walls at extinction. The burner should he operated for the unburned gas injection velocity of around 5 cm/sec. This appears to be a tradeoff between flame stretch at higher injection velocities and heat losses to the burner at lower injection velocities. Shroud nitrogen must be injected at the same velocity as the unburned gas. This is especially important for the determination of the upper flammability limit.

The authors have empirically demonstrated that the wider flammability limits observed by others [e.g., 5] in the tubular burner experiments result from the preheating of unburned gas, rather than from the smaller lateral conductive losses. It follows that the flammability data must be corrected for preheating of unburned mixtures. One such correction technique, which can he applied to inert agents, relies on the calculation of the adiabatic temperature of the limit flames for the measured temperature of the unburned gas. This adiabatic temperature is then used to obtain the new composition (corrected flammability limit) in trial and error computations, for the temperature of the unburned gas of 25 “C.

The operation of the burner involves altering the fuel concentration in the unburned mixture in 1 min time steps until extinction. No repetitive tests are needed to bracket the flammability limits, as are required both by the ISO 10156 and ASTM E681 standards. The experience with the tubular burner in our laboratory indicates that the method is especially useful for scoping studies, which require testing of a large number of refrigerants or fire suppressants.

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